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## **Atmospheric Propagation of THz Radiation**

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# **Atmospheric Propagation of THz Radiation**

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## **Abstract**

In this investigation, we conduct a literature study of the best experimental and theoretical data available for thin and thick atmospheres on THz radiation propagation from 0.1 to 10 THz. We determined that for thick atmospheres no data exists beyond 450 GHz. For thin atmospheres data exists from 0.35 to 1.2 THz. We were successful in using FASE code with the HITRAN database to simulate the THz transmission spectrum for Mauna Kea from 0.1 to 2 THz. Lastly, we successfully measured the THz transmission spectra of laboratory atmospheres at relative humidities of 18 and 27 %. In general, we found that an increase in the water content of the atmosphere led to a decrease in the THz transmission. We identified two potential windows in an Albuquerque atmosphere for THz propagation which were the regions from 1.2 to 1.4 THz and 1.4 to 1.6 THz.



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## Introduction

Terahertz technologies have been proposed as solutions to many current problems such as remote sensing of bio-chem agents and explosives, covert terrestrial and space-based communications, and cold-body tracking. While the promise for terahertz looms large, there are many issues that need to be resolved for full exploitation of this frequency range. Some address the technical immaturity of the technology in this frequency range, including the lack of compact, tunable sources, detectors, and other standard optical components. Besides the hardware issues there are three very fundamental issues: 1) What is the atmospheric transparency in the THz frequency range? 2) How do we separate desired signals from the thermal blackbody-background created by any warm object present in the THz? and 3) What are the signatures of materials between 0.1 and 10 THz?

Rotational transitions of molecules are very efficient absorbers of THz radiation. This makes molecular sensing very attractive, but water vapor absorption lines are extremely ubiquitous and result in relatively short atmospheric propagation paths through most of the THz range. This may actually be of interest for certain applications such as short-range communication where rapid attenuation is desirable. Nevertheless, knowing accurately the location and width of atmospheric transmission windows will be important to facilitate any application in this frequency band.

Background is also a severe issue in this frequency range, much more so than for other frequency ranges due to blackbody radiation. The spectral density of background radiation from any room temperature object increases at the long wavelengths by about four orders of magnitude per decade. Since this is so prevalent, if we wish to get clean signals one has to think about coherent detection, very narrow linewidth sources, and very narrow linewidth detectors, to minimize saturation of the detector. For example if we are trying to track a cold body in space from earth we need to worry about the transmission of the atmosphere and possibly the background from the atmospheric emission. However, the background of space is very low, since it looks like a 3 K background. To track a cold-body from space, we need to know what the background from earth looks like. The earth will emit as a 300 K blackbody, but the atmosphere will absorb most of that, so the question then is what the warm atmosphere actually emits.

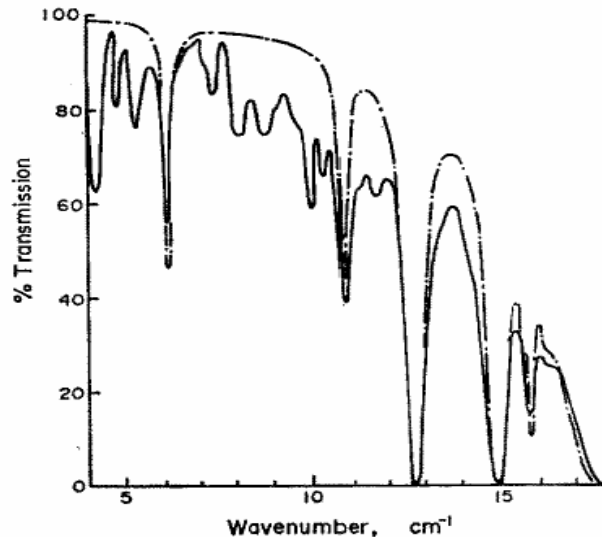
For remote sensing, there may be many frequency ranges that one could exploit. The material of interest may fluoresce when excited with UV light, have vibrational resonances in the MIR, and rotational transitions in the THz. While any one of these measurements may be sufficient in the lab, in a real world environment other materials may also fluoresce at the same wavelengths, or have similar vibrational lines. If the expected signal arises in two or three different wavelength ranges, however, added specificity should be achieved. While the signatures of many molecules are known in the terahertz, previous investigations have not addressed signatures of bio-chem or explosive agents or the discrimination capabilities between these and other molecules.

With these technical problems in hand, THz propagation through the atmosphere is crucial to understand to facilitate any type of application utilizing THz technology. Thus this investigation aims to research the current knowledge base on atmospheric propagation of THz radiation and to conduct preliminary laboratory measurements of Albuquerque atmospheres with varying relative humidities.

## Literature Survey: Best Experimental Atmospheric THz Propagation Data

In our literature search, we wanted to obtain the best experimental data on the atmospheric absorption of THz radiation from 0.1 to 10 THz. With this information, we wanted to identify the unmeasured sections of the THz bandwidth and potential transmission windows for THz propagation.

At sea level, the best experimental measurements we found for horizontal propagation through the atmosphere was work done by R.J. Emery et. al. They conducted experiments from 0.1 to 0.450 THz using an FTIR at 6 GHz resolution. They looked at propagation as a function of temperature, relative humidity (62% to 95%) and path length (83 – 216m). Figure 1 shows an example of their measured atmospheric transmission spectrum.

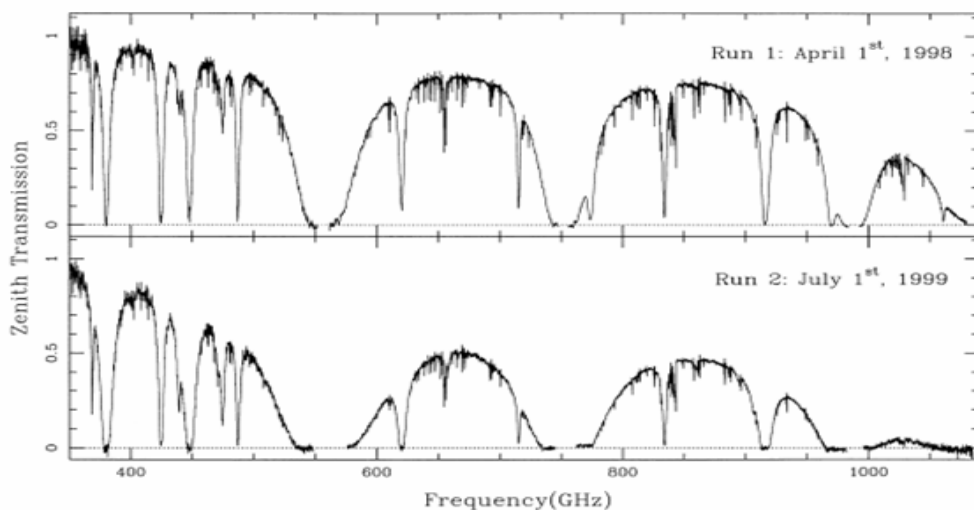


**Figure 1.** Sea level THz transmission spectrum from 0.1 to 0.450 THz. The solid line is the actual measured data and the dash dot line is the calculated transmission spectrum. [R.J. Emery, P. Moffat, R. A. Bohlander and H.A. Gebbie. *Journal of Atmospheric and Terrestrial Physics*, Vol. 37, pp. 587-594 (1975)].

The data showed that at sea level, there are 25 % to 75 % transmission windows from 0.1 to 0.36 THz. In addition, as the relative humidity increased, the windows got narrower and shallower. The shortcomings of this data were: 1) spectral range stops at 0.450 THz 2) window absorptions are larger than theoretical calculations for monomeric water 3) no measurement of dry atmosphere opacity and 4) no pressure broadening studies on water and other atmospheric absorbers. At this time, there is no experimental data on the propagation of THz at sea level atmospheres beyond 450 GHz.

At higher altitudes, the only experimental data available in our frequency range was Naylor's and Pardo's work on Mauna Kea, Hawaii. The measurements are done at a 4.2 km altitude with relative humidities (RH) of 2 % and 12 %. They conducted vertical propagation measurements using Fourier transform spectroscopy with a resolution of 200 MHz from 0.35

to 1.1 THz. They showed that atmospheric propagation of THz radiation at higher altitudes improves dramatically. They found 25 % to 95 % transmission windows from 0.37 to 1.06 THz at 2 % RH and 25 % to 85 % transmission windows from 0.37 to 0.91 THz at 12 % RH. Figure 2 show the best high altitude experimental measurement in our frequency range. At this time, there are no experimental measurements of THz propagation through high altitude atmospheres beyond 1.1 THz.



**Figure 2.** Thin atmosphere THz transmission spectrum in Mauna Kea, Hawaii from 0.35 to 1.1 THz. Top spectrum is at 2 % relative humidity and the bottom spectrum is at 12 % relative humidity. [J. R. Pardo, E. R. Serabyn and J. Cernicharo. *Journal of Quantitative Spectroscopy & Radiative Transfer*, Vol. 68, pp. 419-433 (2001)].

Lastly, we discovered that NASA's Jet Propulsion Laboratories maintains a database from 0 – 10 THz for 298 atoms and molecules that have atmospheric and astrophysical significance. This data was gathered from NASA's microwave limb sounding program. However, we learned that not all the lines in the database are experimentally measured. Instead, they have been calculated given an appropriate Hamiltonian model that describes the molecule.

## Experimental Summary and Transmission Windows from Best Experimental Data

From the literature survey of the best experimental THz propagation data, we put together a summary table of the experimental details of each experiment and the transmission windows derived from the data. See Table 1.



**Table 1.** Summary of experimental THz transmission data from the literature survey.

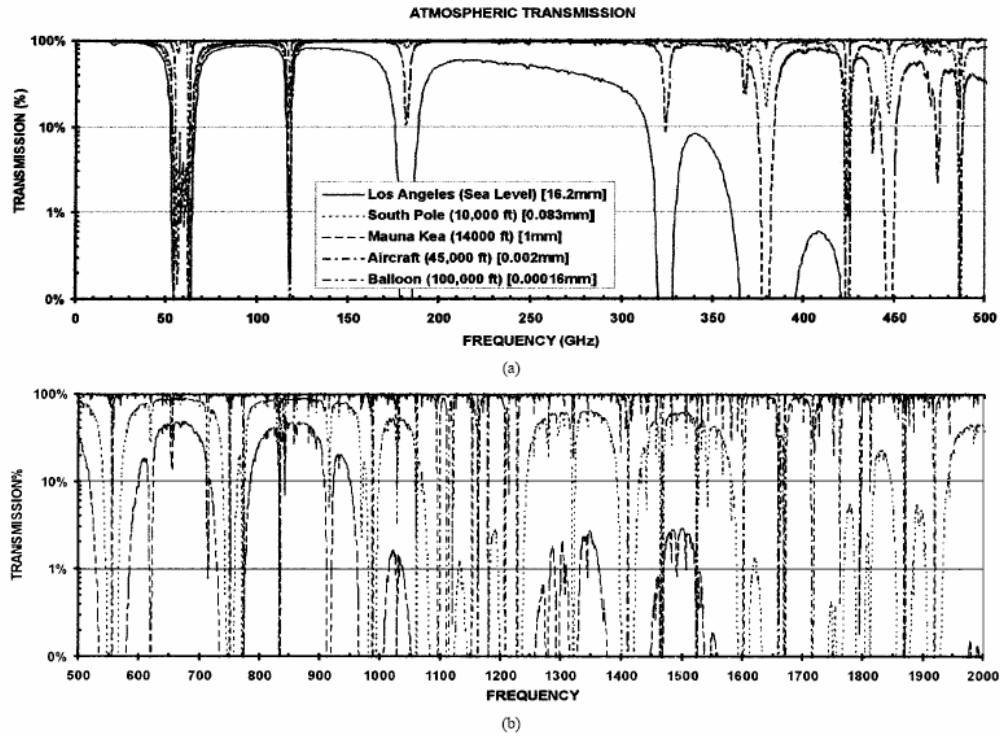
Source	Date	Technique	Resolution (MHz)	Spectral Range Covered (THz)	Location	Altitude Above Sea Level (km)	Ground %RH	Ground Atmos Pressure (mbar)	Ground Temp (K)	>25% Trans Window Width (THz)	Window Depth Trans Range
Emery, R.J. (1975)		FTIR	6000	0.120 - 0.450		0	62	1013	293	0.090 - 0.375	70 - 25
							83 m eter path length horizontal trans			0.390 - 0.444	60 - 25
							0.85mm precipitable water			0.456 - 0.474	30 - 25
Emery, R.J. (1975)		FTIR	6000	0.120 - 0.450		0	95	1013	282	0.120 - 0.186	60 - 25
							216 m eter path length horizontal trans			0.189 - 0.327	50 - 25
							1.85mm precipitable water			0.333 - 0.366	30 - 25
Emery, R.J. (1975)		FTIR	6000	0.120 - 0.450		0	82	1013	275	0.150 - 0.375	60 - 25
							216 m eter path length horizontal trans			0.399 - 0.429	40 - 25
							1mm precipitable water				
Pardo, J.R. (2001)	Jul-99	FTS Zenith Spectra	200	0.35 - 1.2	Mauna Kea	4.1	12	620 465 torr	272 (30F)	0.370 - 0.415	85 - 25
										0.425 - 0.435	70 - 25
										0.455 - 0.475	65 - 25
										0.480 - 0.485	55 - 25
										0.490 - 0.520	50 - 25
										0.625 - 0.712	50 - 25
										0.790 - 0.830	45 - 25
Pardo, J.R. (2001)	Apr-98	FTS Zenith Spectra	200	0.35 - 1.1	Mauna Kea	4.1	2	620	272 (30 F)	0.370 - 0.415	95 - 25
										0.425 - 0.440	90 - 25
										0.455 - 0.485	85 - 25
										0.487 - 0.535	80 - 25
										0.580 - 0.620	65 - 25
										0.625 - 0.712	75 - 25
										0.780 - 0.830	70 - 25
										0.835 - 0.910	75 - 25
										0.920 - 0.970	65 - 25
										1.025 - 1.060	35 - 25
Naylor, D.A. (2000)	May-98	Polarized FTS Zenith Spectra	150	0.35 - 1.1	Mauna Kea	4.1	N/A	625	280	0.370 - 0.415	85 - 25
										0.425 - 0.435	70 - 25
										0.455 - 0.475	65 - 25
										0.480 - 0.485	55 - 25
										0.490 - 0.520	50 - 25
										0.625 - 0.712	50 - 25
										0.790 - 0.830	45 - 25
										0.835 - 0.910	45 - 25

## Literature Survey: Best Theoretical Atmospheric THz Propagation Data

For the theoretical calculations survey, we wanted to understand what were the major codes used to calculate spectra in this region and what were the weaknesses and strengths of these codes.

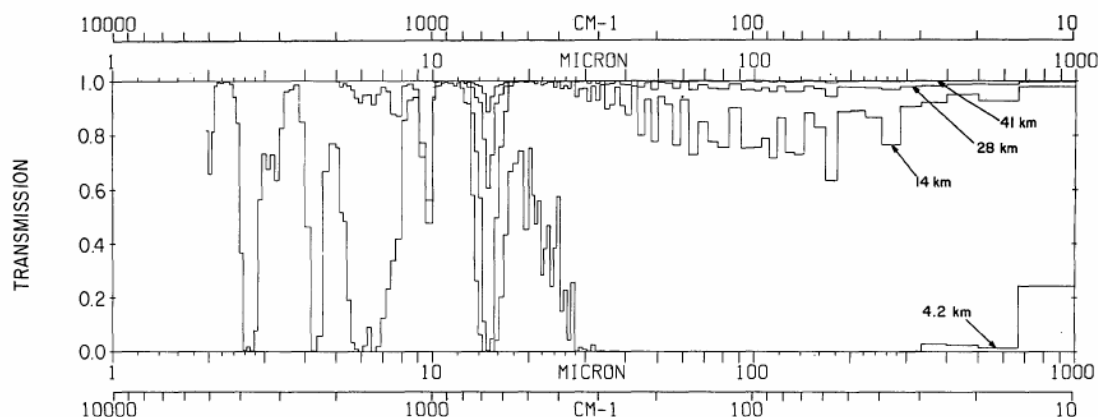
At sea level, Woolard, Siegel and Emery did the best theoretical calculations. Their combined spectral ranges for their calculations were from 0 to 2 THz. The radiation codes used were PcLnWin, Airhead and AFGL. The atmospheric windows lied within the range of

0 to 0.36 THz where the transmission was 25 % – 100 %. Beyond this spectral range the transmission was very small. However, there wasn't an explicit statement about the resolution of these calculations. Figure 3 shows Siegel's' calculated sea level transmission spectrum of the atmosphere. The theoretical models containing resonances beyond 450 GHz are not experimentally validated for thick atmospheres and hence are limited.



**Figure 3.** Atmospheric THz transmission at various locations and altitudes for a given precipitable water vapor pressure (in millimeters) from 0 to 2 THz. [P. H. Siegel. IEEE Transactions on Microwave Theory and Techniques, Vol. 50, pp. 910-928 (2002)].

At higher altitudes, Traub did the most extensive calculations using AFGL code from 0.3 to 12 THz with a 1.5 GHz resolution. He did a slant propagation using a Curtis-Godson approximation for their atmospheric model together with AFCRL database. Figure 4 shows his spectral results. He found that the windows of transmission increase from 25 % to 99 % in this spectral range as you go from 4.2 km to 41 km. As a result, theoretical models containing resonances from 1.1 THz to 10 THz are not experimentally validated for thin atmospheres.



**Figure 4.** High altitude, theoretical calculations for THz transmission from 0.3 to 12 THz. [W.A. Traub and M.T. Stier. *Applied Optics*, Vol.15, No. 2, pp.364-377 (1976)].

For theoretical calculations, the most commonly used models to estimate absorptions in the atmosphere are:

- a) Atmospheric transmission model of Grossman (AT)
- b) Atmospheric transmission at microwaves model of Cernicharo (ATM)
- c) Microwave propagation models of Liebe (MPM89, MPM93)
- d) Other models based on HITRAN, JPL and GEISA line catalogs

All the models with the exception of Liebe's model lack the inclusion of the continuum terms (dry and water vapor opacities) in their calculations. However, the peak resonance information is very accurate from 0 to 1.1 THz. In addition, the H<sub>2</sub>O continuum-like terms defined in MPM models are not accurate in the submillimeter range. For the dry atmosphere opacity term, the models get better if one incorporates N<sub>2</sub>-N<sub>2</sub>, O<sub>2</sub>-N<sub>2</sub>, N<sub>2</sub>-O<sub>2</sub>, and O<sub>2</sub>-O<sub>2</sub> collisions.

## Code Parameter Summary and Transmission Windows from Best Theoretical Data

From the literature survey of the best theoretical THz propagation data, we put together a summary table of the theoretical details of the calculations and the transmission windows from their spectra. See Table 2.

**Table 2.** Summary of theoretical THz transmission data from the literature survey.

Source	Code	Spectral Range Covered (THz)	Altitude Above Sea Level (km)	Ground %RH	Ground Atmos Pressure (torr)	Temp (K)	>25% Trans Window Width (THz)	Window Depth (%Trans Range)
Woolard, D.L. (2003)	PcLnWin Based on Fascode	0 - 1.2	0	60	760	293	0 - 0.060 0.090 - 0.180 0.210 - 0.330 0.335 - 0.360 transmission < 25% at freq > 0.360 THz	25 - 100 25 - 100 25 - 100 25 - 60
Siegel, P. H. (2002)	Airhead Software Erich Grossman	0 - 2.0	0 Los Angeles	16.2 mm precipitable water vapor	760	293	0 - 0.055 0.065 - 0.115 0.120 - 0.170 0.190 - 0.315 transmission < 25% at freq > 0.315THz	25 - 100 25 - 80 25 - 80 25 - 60
Traub, W.A. (1976)	AFGL	0.300 - 15 1.5 GHz resolution	4.2 Mauna Kea	1.2mm precipitable water vapor	450 1mbar = 750mTorr	228	from 300GHz to 12 THz transmission < 25%	
Traub, W.A. (1976)	AFGL	0.300 - 15 1.5 GHz resolution	14 aircraft	2.5 ppmv	106.2	217	from 300GHz to 12 THz transmission is 80%	
Traub, W.A. (1976)	AFGL	0.300 - 15 1.5 GHz resolution	28 balloon	2.5 ppmv	12.15	230	from 300GHz to 12 THz transmission is 95%	
Traub, W.A. (1976)	AFGL	0.300 - 15 1.5 GHz resolution	41 balloon	2.5 ppmv	1.89	268	from 300GHz to 12 THz transmission is 99%	

**Theoretically Calculated Atmospheric Propagation Spectra in the THz Regime**

Source	Code	Resolution (MHz)	Spectral Range Covered (THz)	Propagation Path	Altitude Above Sea Level (km)	Ground Water Vapor Density (g/m <sup>3</sup> )	Ground Atmos Pressure (torr)	Ground Temp (K)	10dB/km Atten. Window Width (THz)	Window Depth (dB/km Range)
Emery, R.J. (1979)	AFGL	N/A	0.1 - 1	Horizontal	0	7.5	760	293	0.100 - 0.180 0.195 - 0.320 0.340 - 0.360 total attenuation > 0.360 THz	0.5 - 10 2 - 10 8 - 10
Emery, R.J. (1979)	AFGL	N/A	0.1 - 1	Vertical	3	7.5	760	293	0.100 - 0.180 0.195 - 0.320 0.340 - 0.360 0.400 - 0.450 0.455 - 0.480 total attenuation > 0.480 THz	0.5 - 10 0.5 - 10 1 - 10 4 - 10 9 - 10
Emery, R.J. (1979)	AFGL	N/A	0.1 - 1	Vertical	12	7.5	760	293	0.100 - 0.120 0.140 - 0.440 0.445 - 0.455 0.460 - 0.480 0.485 - 0.550 0.575 - 0.750 0.770 - 0.790 0.795 - 1.00	0.05 - 10 0.1 - 10 0.5 - 10 0.5 - 10 0.5 - 10 0.5 - 10 0.8 - 10 1 - 10

## Atmospheric Modeling

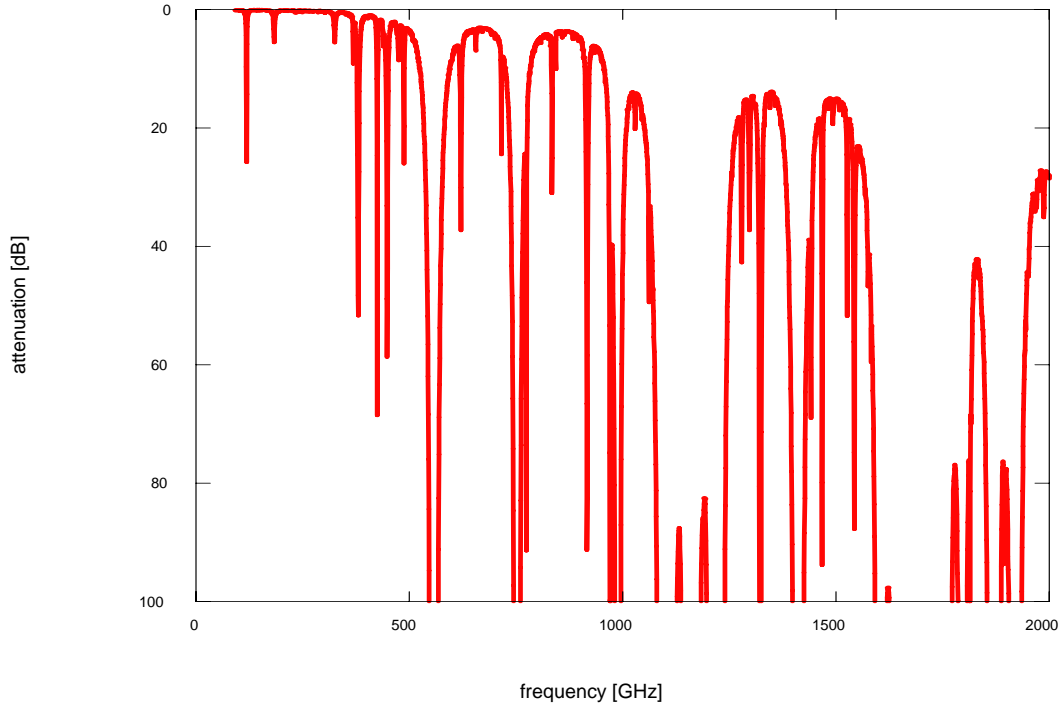
To conduct radiation simulations, three components are required: an atmospheric model, database of spectral information, and radiation code. Radiation codes apply the physical conditions to spectral lines such as Doppler and pressure broadening to the spectral information. The categories of radiation code are line by line, wideband, and narrow band.

Given that there are many radiation codes that are in use for making atmospheric transmission calculations. Of the types mentioned above, the line-by-line codes are the most detailed since they are based on fundamental physics and carry out the numerical calculations to a high degree of accuracy. Briefly, the line-by-line codes assign an absorption coefficient to a sufficiently small frequency interval by summing over all lines the product of the integrated line intensity and the line shape factor centered in the frequency interval for the appropriate temperature and pressure. The absorption coefficient is then used in calculating quantities such as transmission. There are several line-by-line codes available including FASCODE, LBLRTM and FASE (FASCODE for the environment.)

As indicated, in order to use a line-by-line radiation code one needs a spectroscopic database, which contains the necessary parameters to assign line shape as a function of temperature, pressure, and absorption coefficients. There exist several databases for this type of work such as HITRAN spectroscopic database, JPL, GEISA database, and SOA Terahertz Toolbox. However, the most commonly used database is the HITRAN spectroscopic database.

Currently, we have the latest version of FASE, which we are using in conjunction with the latest HITRAN atmospheric molecular spectroscopic database. We have successfully used this combination to model transmission as a function of frequency for the special case of Mauna Kea and have found satisfactory agreement to experimental data for that case. Figure 5 shows our results of Mauna Kea.

In our study, we uncovered four issues with using radiation codes for atmospheric modeling of sea level atmospheres. The first issue is the line shapes of the resonances are poorly understood for atmospheric molecules (specifically water vapor). We don't yet fully understand the extent of the "wings" of water's absorption resonances as well as the pressure broadening effects on these transitions. Secondly, the type of numerical approximations used in calculations affects the result (line shape, linewidth, absorption continuum). Third, interdependency of model, radiation code and database may cause spurious results. Lastly, there are no experimental measurements of thick atmospheres beyond 450GHz to validate the models used in the calculation.



**Figure 5.** Simulation of THz transmission in Mauna Kea from 0.1 to 2 THz using FASE code and HITRAN database.

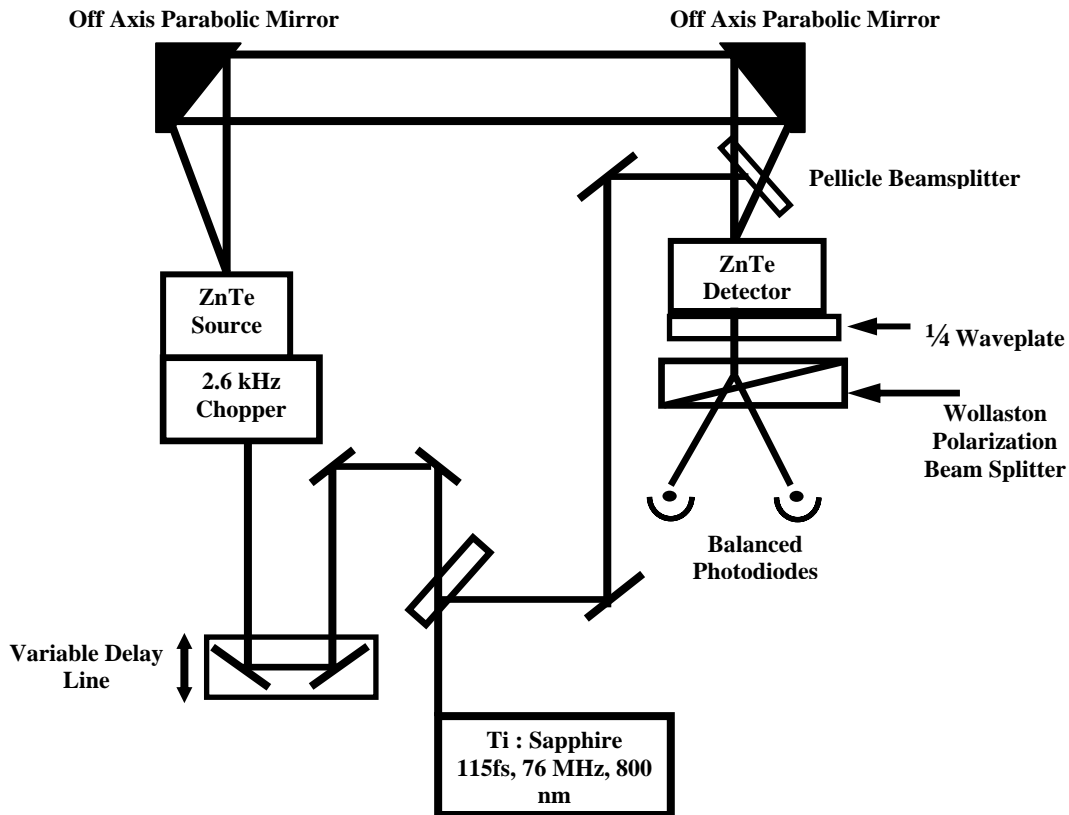
## Experimental Measurements

The experimental measurements we conducted on the propagation of THz radiation through the atmosphere were limited to a one meter path length laboratory atmosphere with two different relative humidities 18 % and 27 %. The goals of this preliminary experiment were to 1) get a benchmark spectrum of an Albuquerque atmosphere 2) determine the effect of relative humidity on THz propagation and 3) identify potential transmission windows. These measurements were made utilizing the THz time domain spectroscopy technique with 8 GHz resolution and a spectral width from 0.6 to 2.6 THz.

## Experimental Details

The atmospheric water vapor spectra were measured with a pulsed, THz-TDS spectrometer utilizing optical rectification and free space, electro-optic sampling for THz generation and detection, respectively. Figure 6 shows a diagram of the ZnTe crystal-based THz-TDS spectrometer used in this investigation. A mode-locked, Ti:Sapphire oscillator (Coherent Mira 900) with a 115 fs pulsewidth, 76 MHz repetition rate and centered at 800 nm is split into a high energy pump and low energy probe beam. The vertically-polarized, pump beam propagates through a delay line (Newport ILS150PP) with 0.5  $\mu$ m resolution and is focused onto a 10 x 10 x 0.9 mm thick <110> ZnTe crystal (eV Products) at normal

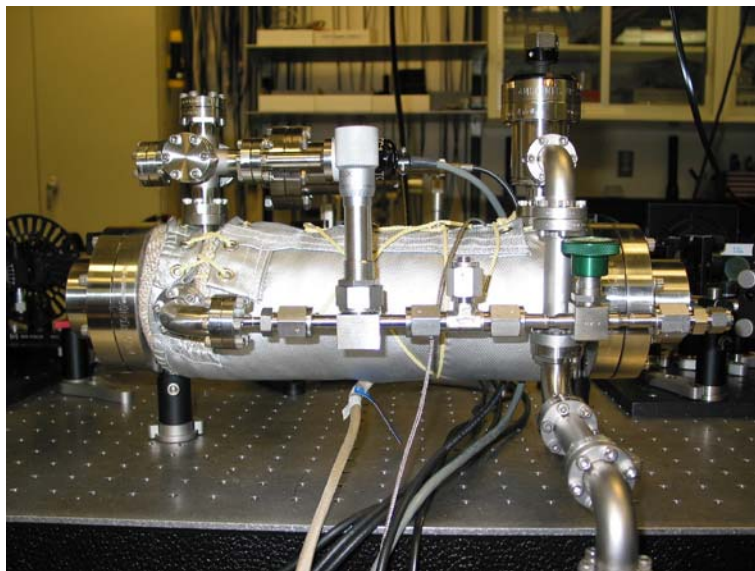
incidence with an average power of 311 mW and a beam diameter of 500  $\mu\text{m}$ . The THz beam that is generated on the output face of the ZnTe crystal is collected and collimated by a 10 cm focal length, off-axis parabolic mirror with a 90 degree reflection angle. The THz beam traverses a path length of 94 cm and is focused onto a 10 x 10 x 0.9 mm thick <110> ZnTe crystal sensor. The low energy probe beam is vertically polarized and is focused to a beam diameter of 50  $\mu\text{m}$  as it co-propagates with the THz beam through the ZnTe crystal. The probe beam then experiences a polarization rotation directly proportional to the THz electric field strength. The polarization change is analyzed by allowing the probe beam to traverse through a  $\frac{1}{4}$  waveplate oriented at 45 degrees. The polarization components are split by a Wollaston prism with a 10 degree divergence angle and the resulting split beam is weakly focused onto a large area balanced photoreceiver (New Focus Model 2307) with a gain setting of  $10^5$  V/A. The average probe optical power of each polarization component present at the balanced photodiode is 3.8 mW. The THz generation is modulated at 2.6 KHz and the differential signal is measured by a SRS Model 830 DSP lock in amplifier with an integration time of 300 ms and a low pass filter roll off of 12dB/Oct. The data acquisition is controlled by LabVIEW 6.0 on a desktop PC. In addition, the complete propagation path of the THz beam was enclosed in a nitrogen purged environment to prevent the interaction of atmospheric water vapor with the THz beam during our reference scans.



**Figure 6** Terahertz time-domain spectroscopy spectrometer overview.

## Gas Cell Design

Figure 7 shows a gas cell we designed during this project to conduct atmospheric measurements as well as follow-on measurements of other materials of interest using THz-TDS. This cell was designed with 5 mm thick z-cut crystal quartz windows with a two-inch diameter. The propagation length of the cell was 38 cm and the cell can be heated to 200 C. For the atmospheric study, we measured ambient atmosphere encased in the volume of the gas cell and within the nitrogen purge box.



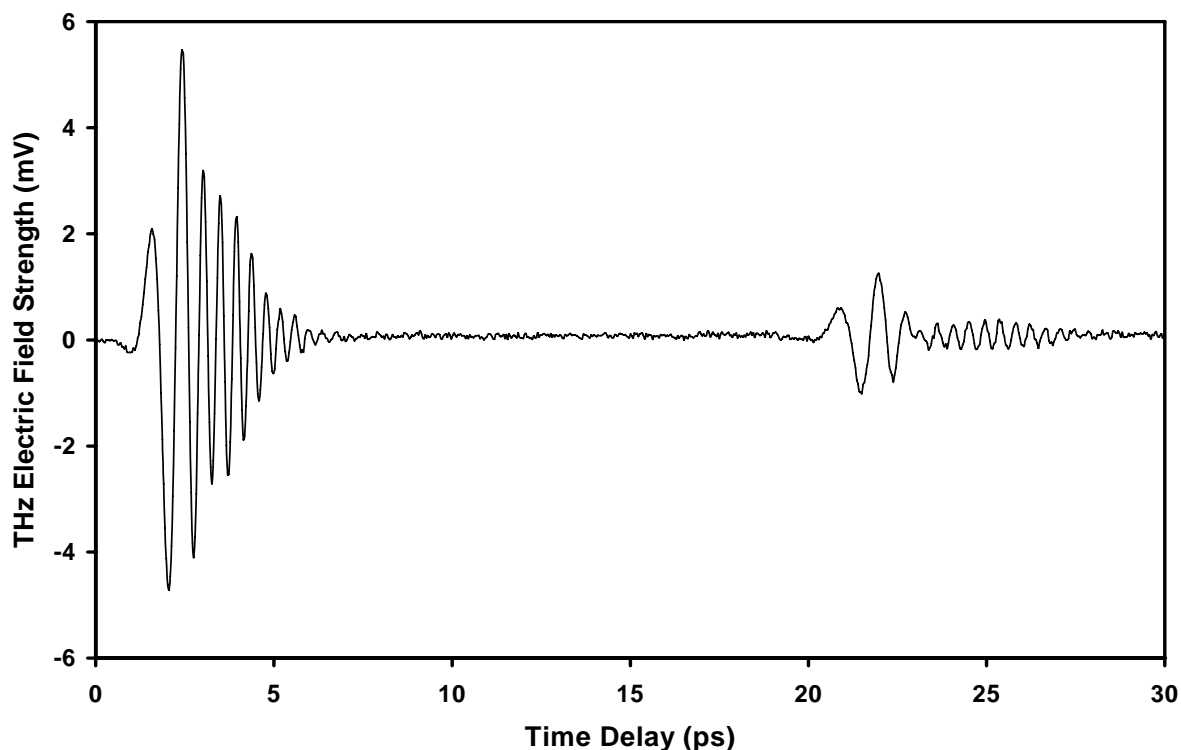
**Figure 7.** Photo of the gas cell we designed to conduct atmospheric vapor measurements.

## Atmospheric Water Vapor Measurements at Different Relative Humidities

### Results

The experimental resolution for the measurements of atmospheric water vapor transitions is limited by the pressure broadening of the lines. Given that the atmospheric pressure in Albuquerque, New Mexico is 620 Torr and assuming a standard pressure broadening coefficient of 10 MHz / Torr, the pressure-broadening-limited resolution is 6.2 GHz. However, we chose to have an experimental resolution of 8 GHz. Therefore, our experimental scan lengths are truncated at a 125 ps in the time-domain. Figure 8 shows an example of the time-domain profile of our *reference* scan.

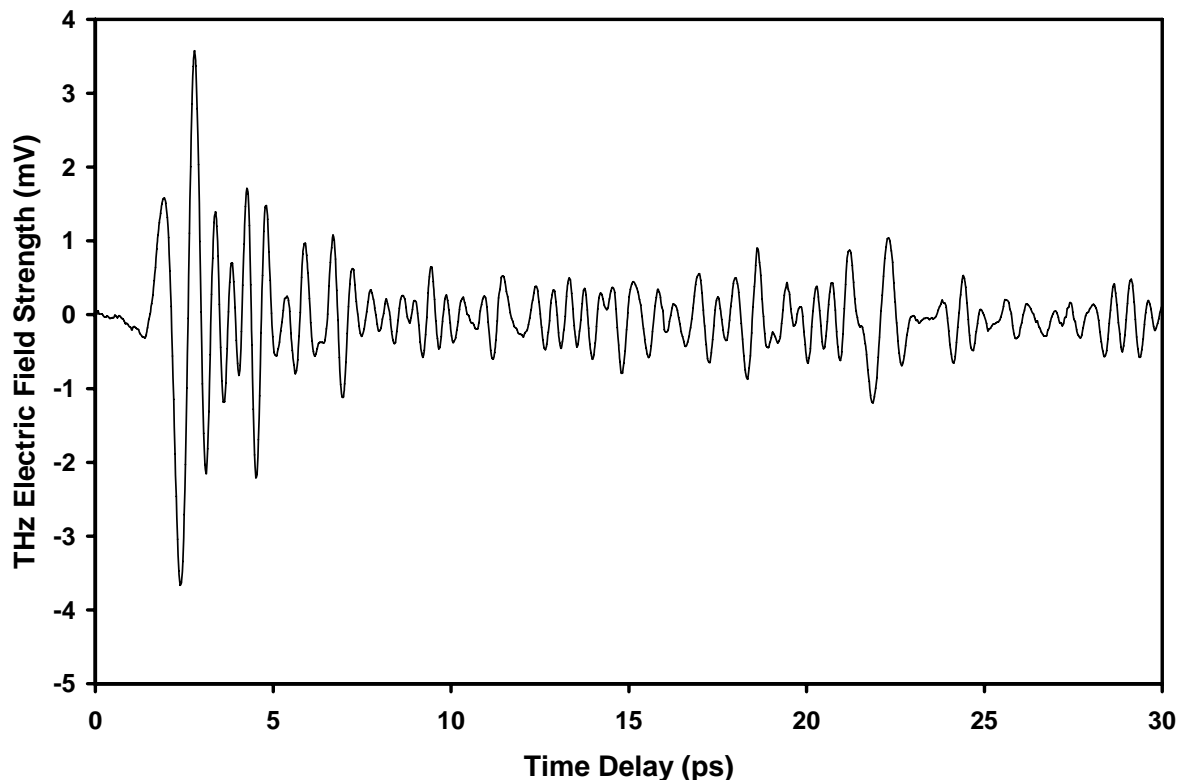




**Figure 8.** A typical time-domain profile of a nitrogen-rich environment with a relative humidity of  $0 \pm 2\%$  at  $23.4\text{ }^{\circ}\text{C}$ . This reference scan extends out to 125 ps, but only 30 ps are shown. No structure appears from 7 to 18 ps indicating no THz absorption or dispersion by water vapor molecules.

In this scan, the THz radiation is propagating through a nitrogen-rich environment with a relative humidity of  $0 \pm 2\%$  as measured with a hygrometer (Dickson TM325). The profile clearly shows a main THz pulse from 0 – 5 ps with a peak amplitude of approximately 6 mV and a secondary pulse with a peak at 20.5 ps. The peak at 20.5 ps is the weaker reflected pulse of the main THz beam as it propagated through two 0.9 mm thick ZnTe crystals. These reflections are reproducible and divide out in frequency space when we normalize the reference scan to the water vapor scan in our analysis. Also in Figure 8, note the signal directly after the main pulse (7 to 18 ps) shows a flat line devoid of structure or modulation which confirms that there is no interaction between the THz radiation and water vapor in the reference scan.

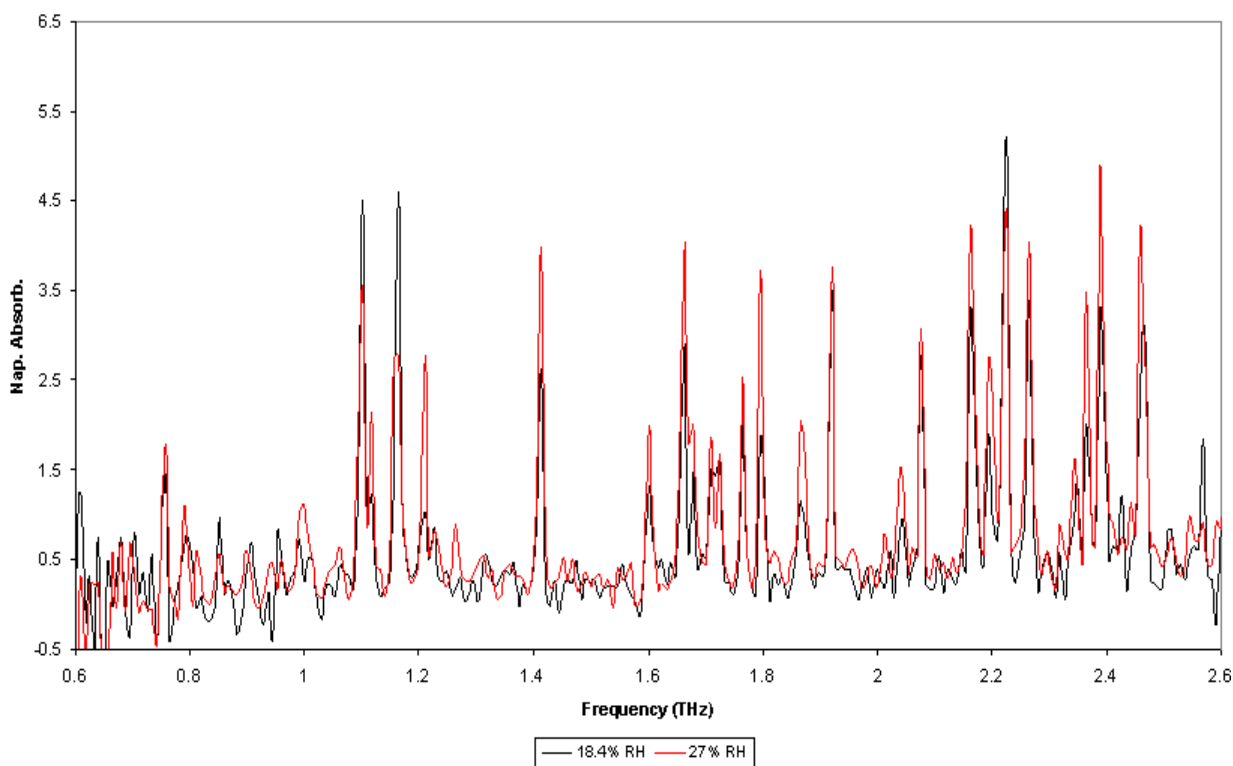
The water vapor scan was taken by shutting off the nitrogen purge and exposing the THz beam to the atmosphere as well as filling the gas cell with atmosphere. We measured two absorption spectra for atmospheric water at two different times of the year. Both spectra were measured at a temperature of  $23.4\text{ }^{\circ}\text{C}$ , with relative humidities of  $18.4 \pm 2\%$  and  $27 \pm 2\%$ . Figure 9 shows the time-domain profile for atmospheric water vapor with a relative humidity of  $27 \pm 2\%$ .



**Figure 9.** A time-domain profile of atmospheric water vapor with  $28 \pm 2\%$  relative humidity at  $23.4^\circ\text{C}$ . This sample scan extends out to 125 ps, but only 30 ps are shown. Notice how the time-domain profile of the water vapor shows a dramatic change in the shape of the waveform due to the strong absorptions and dispersion of the THz radiation by water vapor molecules.

One can clearly see not only the reduction in the main pulse peak amplitude from approximately 6 mV to 3.5 mV when compared to the reference, but also the modulation of the THz waveform due to the absorption and dispersion of the THz beam when it interacted with water vapor in the 5 to 20 ps time frame. This occurs because the water vapor is creating phase and amplitude changes in the spectral components of the THz bandwidth that constructively and destructively interfere to produce a resultant waveform different from the reference.

To obtain the absorption spectrum of the atmospheric water vapor, a FFT of the time-domain profiles for the reference and the sample scans was calculated. Since absorbance calculations are intensity based expressions, the resulting spectral amplitude profiles were squared to produce a power spectrum for both the reference and the water vapor scans. Finally, the Napierian absorbances were determined by taking the natural log of the ratio between the reference scan spectral power and the water vapor scan spectral power. Figure 10 shows the resulting absorption spectrum of atmospheric water vapor from 0.6 to 2.6 THz.



**Figure 10.** Measured absorption spectra of atmospheric water vapor. The atmospheric pressure is 620 Torr with a relative humidity of  $18.4 \pm 2$  % (black) and  $27 \pm 2$  % (red) at  $23.4^{\circ}\text{C}$ . The propagation length of the THz beam through the atmosphere is 94 cm.

## Discussion

In order to measure the rotational absorption resonances of any molecule utilizing THz-TDS, the molecule must have a permanent dipole moment. Out of the most abundant molecules in the atmosphere which are diatomic nitrogen (78.08 %), diatomic oxygen (20.95 %), water vapor (0.03 %), atomic argon (0.009 %) and carbon dioxide (0.0003 %), only water possesses a permanent dipole [1]. Since water is a bent, asymmetric top rotor with a  $C_{2v}$  symmetry point group, it has a permanent dipole moment with a magnitude of approximately two Debye [2]. Diatomic nitrogen, diatomic oxygen and carbon dioxide will not produce any rotational spectra because their linear symmetry prevents them from having a permanent dipole. Argon atoms, on the other hand, lack a moment of inertia and hence don't have any rotational motion that could produce rotational absorbances. By conducting pulsed, THz-TDS, the experiment itself acts as a filter to isolate the investigation of only the water vapor content in the atmosphere. However, not only will  $\text{H}_2^{16}\text{O}$  be THz active, but also water's isotopes. The relative abundances of the isotopic water species in the atmosphere are approximately  $\text{H}_2^{17}\text{O}$  (0.03 %),  $\text{H}_2^{18}\text{O}$  (0.20 %),  $\text{HD}^{16}\text{O}$  (0.0149 %) and  $\text{D}_2\text{O}$  (0.0000022 %) [3]. In our investigation, it is possible to observe all these isotopes with the exception of  $\text{D}_2\text{O}$  due to our spectrometer's detection limit of 3 ppm.

To determine the vibrational state we were making our rotational line measurements, we had to conduct an analysis on the available spectral and thermal energies in our experiment and compare it to water's fundamental frequencies. The fundamental frequencies for the three vibrational modes of water in the ground electronic state  $X^1A_1$  are  $\nu_1 = 3657 \text{ cm}^{-1}$  (109.7 THz),  $\nu_2 = 1594.7 \text{ cm}^{-1}$  (47.9 THz) and  $\nu_3 = 3755.7 \text{ cm}^{-1}$  (112.7 THz) [4]. In our pulsed THz-TDS experiment we have a spectral range of 0.4 to 2.7 THz and an experimental temperature of 23.4 °C. Since our spectrometer's spectral range doesn't reach any of the fundamental vibrational modes and our translational energy at 23.4 °C is  $308 \text{ cm}^{-1}$ , we are restricted in making absorption measurements of rotational lines in the ground electronic ( $X^1A_1$ ), ground vibrational state (000) of water and its isotopes. This analysis describes that generally, most of the water lines measured are populated in the ground vibrational state. However, due to the complexity of the atmospheric constituents, there may be some excited vibrational states of water as well as magnetic dipole allowed transitions of diatomic oxygen. The complete analysis of the atmospheric spectrum is much too complex and beyond the scope of this paper.

The strongest transitions in our THz absorption measurement belong to water vapor. These lines were validated using JPL's database of water lines. All of our experimental lines agreed with the database within the 8 GHz resolution of our experiment. In terms of the affect relative humidity has with THz absorption, in general it appears as we increase the relative humidity, we get more THz absorption and hence larger water peaks in our spectrum depicted in red.

From the spectrum, we can identify some potential transmission windows. Two areas that show the most promise are 1) 1.2 to 1.4 THz and 2) 1.4 to 1.6 THz. These areas seem to show very little window absorption as well as it is devoid of water structure.

## Conclusions

From our investigation, we found that for sea level atmospheres, transmission windows exist only below 450 GHz and no experimental data exists at sea level beyond 450 GHz. At higher altitudes the THz transmission is greater, but no experimental measurements exist for high altitude atmospheres from 1.1 to 10 THz.

We validated the FASE code successfully by comparing our results with the data measured at Mauna Kea. However, we have difficulty in modeling thick atmospheres beyond 450 GHz because there is no existing experimental data to feed into the radiation code about the linewidths profiles of water vapor.

We built gas cell capable of handling vapors from solids, liquids, and gases up to 200 C. In addition, we measured the THz absorption spectra for water vapor at 18.7 % and 27 % relative humidities. In general, the higher the humidity the larger absorption for most of the states in water vapor. Lastly, we identified two potential windows in the atmosphere for propagation which were the regions from 1.2 to 1.4 THz and 1.4 to 1.6 THz.

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